

ESR Modes in CsCuCl₃ in Pulsed Magnetic Fields

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We present ESR results for 35 – 134GHz in the antiferromagnet CsCuCl₃ at $T = 1.5\text{K}$. The external field is applied perpendicular to the hexagonal c -axis. With our pulsed field facility we reach 50T an unprecedented field for low temperature ESR. We observe strong resonances up to fields close to the ferromagnetic region of $\approx 30\text{T}$. These results are discussed in a model for antiferromagnetic modes in a two-dimensional frustrated triangular spin system.

CsCuCl₃ belongs to the hexagonal ABX₃ type compounds in which linear chains of face sharing octahedra (BX₃) are formed along the c -axis. A strong intrachain ferromagnetic coupling and smaller interchain antiferromagnetic interaction give rise to a triangular spin arrangement in the c -plane below $T_N = 10.5\text{K}$. A cooperative Jahn-Teller transition at 423K distorts the CuCl₆ octahedra [1], but the structure remains hexagonal with a tripled c -axis unit cell. The low symmetry of the local structure leads to the antisymmetric Dzyaloshinsky - Moriya interaction along the chains and gives a helical spin structure along the c -axis with a long period [2].

More recently the research for this compound focussed on two topics: On the one hand an interpretation of the beautiful high field magnetization experiments [3] using quantum fluctuations in this frustrated spin system for $B \parallel c$ [4] and for $B \perp c$ [5] has been given. On the other hand a detailed investigation of the magnetic phases near T_N [6] elucidating the chiral XY state has been carried out.

ESR experiments have been performed for both $B \parallel c$ and for $B \perp c$ in the frequency range 35 – 280GHz [7] and 90 – 380GHz [8] for fields up to 14T in the low temperature range $T < T_N$. In the case $B \parallel c$ one observes the two branches of the antiferromagnetic resonance modes which can be calculated for $B < B_c$ taking the helical spin structure into account [9]. At $B = B_c$ (12.5T at 1.1K) there is a small jump in the magnetization curve [3] and a shift of the upper resonance frequency [8]. B_c is the critical field where the umbrella type spin structure changes to a coplanar structure [4].

For $B \perp c$ the physics is more complicated since the magnetic field breaks the axial symmetry of the system. The helical spin structure for $B = 0$ becomes a distorted helix for $B > 0$. It is possible to get from this incommensurate structure to a commensurate one at a critical field estimated theoretically $B_{c1} = 10 - 11\text{T}$ [5] (but see below). Inclusion of quantum fluctuations on the commensurate state establishes in the magnetization a small plateau at B_{c1} as observed experimentally [3]. The low temperature ESR experiments show again two antiferromagnetic resonance branches with the lower one tending to zero at B_{c1} [7]. Apart from strong resonances we observe also smaller ones in the vicinity of B_{c1} [7,8]. The interpretation of these results is more complicated than in the $B \parallel c$ case. So far simple models like a spinwave calculation for the triangular frustrated planar spins [10] or a classical resonance calculation for this system [7] have been performed.

We performed ESR experiments in very high magnetic fields up to 50T for the $B \perp c$ configuration (For the $B \parallel c$ geometry the resonance frequencies are smaller than 35GHz in the high field region and therefore not attainable for us). We used the pulsed field facility of the Frankfurt High Magnetic Field Laboratory [11] to obtain a maximum field of 50T with a typical pulse length of 30ms. We detect ESR lines in the frequency range 35 – 134GHz with a liquid Helium cooled fast InSb hot electron bolometer. The microwave radiation was generated by Gunn oscillators and Impatt diodes with frequency multipliers. The radiation is transmitted twice through the specimen by reflecting on a Al-coated ceramic disk below the specimen and brought to the bolometer with directional couplers. Pulsed field ESR has been performed before in the pioneering work of Date *et al.* [12].

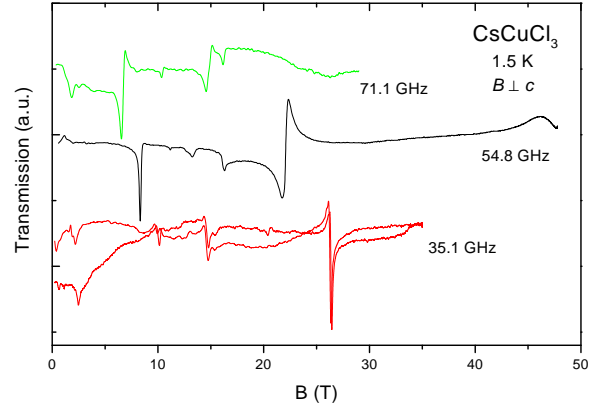


FIG. 1. Transmission of the ESR signal as a function of magnetic field B for three different frequencies at $T = 1.5\text{K}$. The strong resonances exhibit large dispersive effects.

In fig.1 we show typical ESR signals for 35.1, 54.8 and 71.1GHz at 1.5K in the field range up to 48T. The signal amplitude is plotted for the rising section of the field pulse. The resulting thermal effects due to eddy currents of the sample holder or due to magnetocaloric effects in the sample are almost negligible [11] as seen from the 35GHz curve where we show the result for the ascending and descending section of the magnet pulse. From the temperature dependence of the resonances to be discussed below we expect at most a heating effect of $< 0.5\text{K}$ for a 50T pulse. For each frequency we observe two large resonances (e.g. at 8T and 22T for the 54.8GHz result) and up to three smaller resonances (e.g. at

11T, 13T and 16T for the 54.8GHz resonance) which correspond to the small resonances mentioned above.

In fig.2 we plot the observed resonances as a function of magnetic field. The full symbols indicate the strong resonances, the crosses the small ones and the open circles resonances from ref. [7]. In the region of overlap for the data of ref. [7] and our new one the agreement is very good. As to the smaller resonances they partly agree with older results. We believe that these smaller resonances depend on the actual domain state and sample quality. They can also arise as excitations from the complicated ground state especially in the region of the incommensurate - commensurate transition and are very difficult to calculate. In the following we discuss only the large resonances. In fields up to 14T they have been observed already by different groups and in different crystals. A remarkable aspect of fig.2 is the absence of resonances for fields $B > 30$ T for the frequency range 35 – 134GHz. This is shown e.g. in fig.1 for one frequency 54.8GHz.

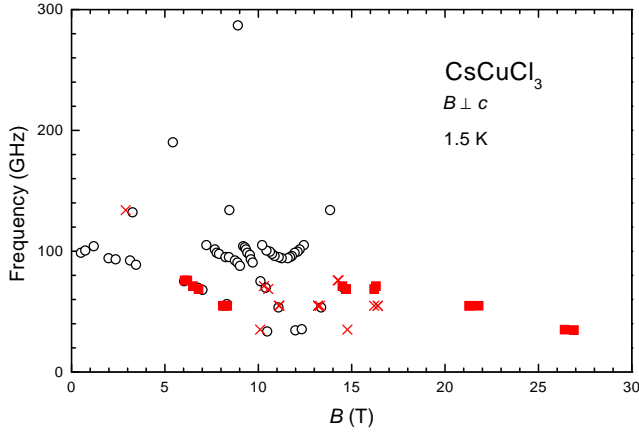


FIG. 2. Frequency versus magnetic field B for $B \perp c$ at $T = 1.5$ K. open circles: results from ref. [7], crosses: small resonances, closed squares: strong resonances from this work. Note the good agreement with former results in the region of overlap.

The calculation of these resonances for the field configuration $B \perp c$ is a very difficult problem. The evaluation of the ground state spin arrangement is already complicated as discussed above. Even in molecular field approximation this is a nontrivial problem, even more so with the important inclusion of quantum fluctuations in the commensurate and incommensurate state [5]. For the discussion of our resonance results we therefore use simplified models.

There are two models which have been discussed and applied to the present state, a spinwave model [10] and a classical calculation of the antiferromagnetic resonance modes [7] (i.e. a $q = 0$ spinwave calculation). Both models assume a two-dimensional triangular frustrated Heisenberg spin model, an approximation which neglects the spiral spin chains along the c -axis. A strong uniaxial anisotropy field of $B_a \approx 0.36$ T confines the spins to the hexagonal plane. Since these models give the salient features of ESR experiments performed up to $B < 14$ T [7] and since the magnetization results [3] give

also the same three regions of spin arrangements discussed in these models ($0 < B < B_{c1}$, $B_{c1} < B < 3B_{c1}$, $3B_{c1} < B$) we would like to know how these calculations describe our results at higher fields. In fig.3 we show the results for the antiferromagnetic resonance model (with $B_{ex} = B_{c1} = 10$ T, $B_a = 0.36$ T) together with experimental results from fig.2 including only the strong resonances. The equations for the antiferromagnetic resonance for the lower mode are:

$$\omega = \gamma \sqrt{3B_a B_{ex} - B^2 \frac{B_a}{B_{ex}} - 2B_a B} \quad \text{for } B < B_{c1}$$

$$\omega = \gamma \sqrt{3B_a B_{ex} + \frac{B^2}{2} - \sqrt{I}} \quad \text{for } B_{c1} < B < B_{c2}$$

$$\omega = \gamma \sqrt{B^2 + 2BB_a - 6BB_{ex} - 6B_a B_{ex} + 9B_{ex}^2} \quad \text{for } B > B_{c2}$$

where γ is the gyromagnetic ratio and

$$I = B^4 \left(\frac{1}{4} + \frac{B_a^2}{2B_{ex}^2} + \frac{3B_a}{4B_{ex}} \right) - B^2 \left(5B_a^2 + \frac{9B_{ex}B_a}{2} \right) + \frac{27B_{ex}^2 B_a^2}{2} + \frac{27B_{ex}^3 B_a}{4}$$

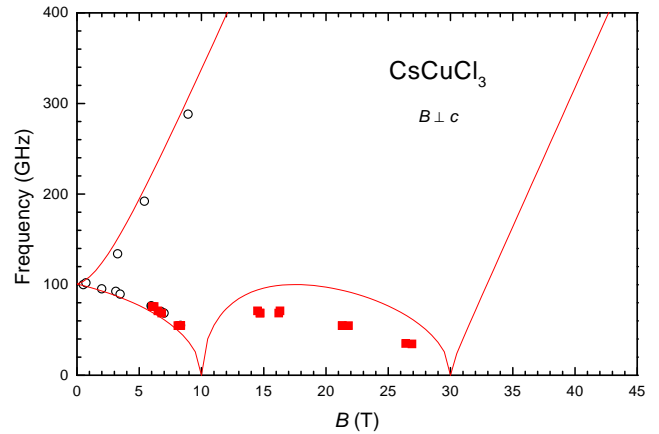


FIG. 3. Fit of the strong resonances from fig.2 with the antiferromagnetic resonance model (full line) as discussed in the text.

Whereas the resonances are well described below B_{c1} by this model as already shown in ref. [7] the agreement is only qualitative for $B > B_{c1} = 10$ T. In addition no resonance has been observed in the saturated region for $B > B_{c2} = 3B_{c1} = 30$ T.

For this latter effect one of the explanations could be the following polarisation features of our ESR experiment. In the high field region we have a ferromagnetic state and the resonance field has to be a transverse field. In our resonance arrangement we have a Faraday geometry but with wavelengths of the order of the tube diameter. Therefore we have cavity modes with the microwave fields b_{rf} parallel and perpendicular to the applied field. Our crystal was placed in a position with predominantly parallel b_{rf} field. Therefore the ab-

sence of transverse resonances for $B > 30\text{T}$ in this frequency regime follows.

We also investigated the temperature dependence of the resonance frequencies. As an example we investigated the 35GHz resonance in the temperature region 1.5 – 3.8K. We find a strong decrease of the resonance field. On going from 1.5 to 3.8K the field changes from 26.4 to 24.8T for $B_{c1} < B < B_{c2}$. This follows from the decrease of the exchange and anisotropy fields with rising temperature.

Recent neutron diffraction experiments [13] demonstrate a transition from incommensurate to commensurate spin structure at 17T for 4.2K and not at B_{c1} as expected before. This result gives support for our model assumption for quasi two-dimensional spin arrangements for $B > 17\text{T}$. A theory for the excitations including Dzyaloshinsky – Moriya interaction and interplanar exchange has not been performed yet.

In conclusion we have demonstrated the feasibility of ESR experiments in pulsed fields up to 50T in CsCuCl_3 at low temperatures. We found resonances in the field region where the triangular spin arrangement is collapsed into partly collinear spins ($B > B_{c1}$) and that these resonances show a softening towards higher fields where the spins ultimately align to a ferromagnetic arrangement. However the resonance frequencies lie appreciably below our calculated modes for a frustrated two-dimensional triangular lattice. We hope that these high field ESR data will stimulate further theoretical investigations of this high field phase and its excitations.

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